Determination of Arsenic (As) Concentration in Different Varieties of Bangladeshi Rice Using Neutron Activation Analysis (NAA)

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Abstract

Rice is the most well known cereal and staple food which serves as major carbohydrate for more than half of the world population. The goal of this study is to determine the selected toxic element (As) in rice samples. Bangladesh is a developing country and the industrial sector of the country is gradually expanding. At the same time it is a threat to the environment. For this experiment, eighteen samples of three common rice were selected namely BRRI-28, BRRI-29 & Miniket. All the samples were analyzed by using Neutron Activation Analysis (NAA) under the irradiation and counting facilities available at the Institute of Nuclear Science and Technology (INST), Atomic Energy Research Establishment (AERE), Savar, Dhaka in Bangladesh. NAA methods were validated by certified reference materials (IAEA-Soil-7, IAEA-SL-1 and Whey powder IAEA-155). The concentration of arsenic (As) was ranged from 0-0.2 \pm 0.003 mg kg⁻¹. The results expressed that the rice samples accumulate the element at different concentrations. Toxic element like As found in alarming level in some samples.

Keywords: Rice; NAA; Tangail; Toxic element

1. Introduction

Rice (Oryza sativa) is the staple sustenance of around 135 million individuals of Bangladesh. It gives Carbohydrate about (75-85) % (Ayasekera & Freitas 2005). It is one of the important foodstuffs worldwide and consumed by half of the world's population (Costa *et al.* 2016; D'Ilio *et al.* 2002; Welna *et al.* 2015; Mahender *et al.* 2016; Diyabalanage *et al.* 2016). Rice plays a vital role in the livelihood of the people of Bangladesh. Recently, concern has been raised about possible contamination of rice worldwide by heavy metals which more acute in case of Bangladesh. The wellsprings of substantial metal in plants are their developers, media like air, soil, supplements, thus on from which over powering metals are taken up by the roots or foliage (Lin *et al.* 2002; Donisa *et al.* 2000). Anthropogenic exercises like industrialization have been adding to the spread of poisonous chemicals into the earth.

There are numerous analytical techniques capable of determining multi-element in foods such as inductively coupled plasma atomic emission spectrometry (ICP-AES), inductively coupled plasma mass spectrometry (ICP-MS), graphite furnace atomic absorption spectrometry (GF-AAS), cold vapor atomic absorption spectrometer (CV-AAS) and Neutron Activation Analysis (NAA) (Parengama *et al.* 2010). Neutron activation analysis is a suitable method to determine elements in plant, soil and food samples (Huynh *et al.* 1997). It is non-destructive, sensitive, high accurate, less sample preparation and multi-element detection method (Huynh *et al.* 1997; Parengama *et al.* 2010). The present study was conducted to assess arsenic (As) concentration in BRRI -28, BRRI -29 and Miniket rice, consumed by the greater portion of Bangladeshi people using NAA. Transfer of As from the soil to the rice and assessment of potential health risk of this metal to human by consumption of rice was also calculated accordingly.

2. Materials and method

2.1 sampling areas

Eighteen rice samples collected from Tangail Sadar Upazilla. This area is located in the Dhaka division near the capital of Bangladesh. Samples were collected from six different sampling areas. The six different sampling areas are shown in Figure 1.

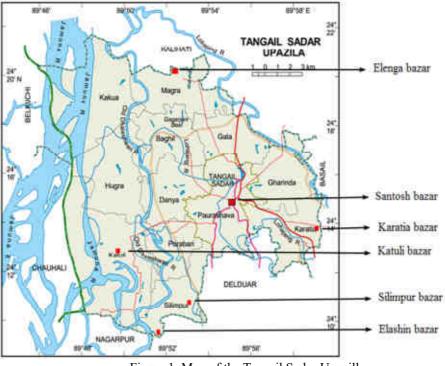


Figure 1. Map of the Tangail Sadar Upazilla

2.2 Sample collection

The samples were collected in the polyethylene bag which are commonly used in our country for staple food. The samples were marked separately by giving the identification (ID) number carefully. To avoid contamination, hand gloves were used to collect the samples. Sample ID and names of the location are shown in Table 1.

Rice variety	Sample identity	Collected from	
	SP-02	Elashin bazar, Tangail	
BRRI-28	SP-05	Silimpur bazar, Tangail	
DKKI-20	SP-08	Elenga bazar, Tangail	
	SP-11	Katuli bazar, Tangail	
	SP-14	Santosh bazar, Tangail	
	SP-17	Karatia bazar, Tangail	
	SP-03	Elashin bazar, Tangail	
BRRI-29	SP-06	Silimpur bazar, Tangail	
DKKI-29	SP-09	Elenga bazar, Tangail	
	SP-12	Katuli bazar,Tangail	
	SP-15	Santosh bazar, Tangail	
	SP-18	Karatia bazar,Tangail	
	SP-01	Elashin bazar,Tangail	
Miniket	SP-04	Silimpur bazar, Tangail	
	SP-07	Elenga bazar, Tangail	
	SP-10	Katuli bazar,Tangail	
	SP-13	Santosh bazar, Tangail	
	SP-16	Karatia bazar, Tangail	

Table 1. Identification of the rice samples

2.3 Sample preparation

18 samples were washed properly by DI (De-ionized) water. It is more pure than distil water. After washing the samples were put into the oven to be dried. The identification number (ID) was given on each Petri dish according to the sample ID. Collected rice samples were put into the Petri dish corresponding to the given ID. The Petri dish containing the rice samples was then allowed to dry in an electric oven at 80° C-90°C until having constant weight. For making small grain size each of the samples was grounded with an agate mortar and pestle. For weighing the rice samples firstly, the weight of each empty Petri dish was set to zero by a digital electronic micro balance. After that, each of the samples was taken into the Petri dishes. The weight of each sample is kept around 300 mg and net weight was recorded in a register book. After weighing the rice samples were made an individual packet with individual identification numbers. The size and shape of packets were kept approximately same. The packets were then preserved, carefully for neutron irradiation. Three standards: IAEA-Soil-7, IAEA-SL-1 and Whey powder were used for this experiment. Nearly 50mg of each standard were prepared in the same way as the sample.

a. Irradiation

The irradiation facility is the Pneumatic Transfer System Rabbit of 3 MW TRIGA Mark-II research reactor, AERE, Savar, Dhaka. The reactor power is 2.4 MW. In the process of long irradiation the total number of sample and standards were taken into a long polyethylene irradiation tube at a time; then it was kept close tightly and was sent for irradiation with the ~ 6.08×10^{13} ncm⁻²sec⁻¹ thermal neutron flux for 7 minutes. After long radiator samples were turned into highly radioactive. For this reason, they usually were not handled immediately. The properties of used radionuclide, γ -energies and other details of the analysis are presented in Table 2.

Table 2. Nuclear	parameters of interested element.
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Element	Radionuclide	Half-life(sec)	γ-ray(keV)	Irradiation time(min)
As	⁷⁶ As	94680	559.1	7

2.5 Gamma ray counting

Gamma ray counting was performed for irradiated samples and standards using the HPGe detector, CANBERRA [25% relative efficiency, 1.85 KeV resolutions] equipped with PC based ORTEC DSpec Digital gamma spectrometry system with the following conditions. For irradiated samples a total of 2 counting were performed:

I. 1st counting: counted for 30 min. after 2 days of decay time.
II. 2nd counting: counted for 120 min. after 20 days of decay time.

2.6 Spectrum analysis

Gamma peak analysis was performed using the program Hyper met PC and also checked each spectrum manually.

2.7 Concentration Calculation

The concentration calculation equation is given below which calculate the concentrations of different elements was carried out based on the relative standardization approach. The activation equation for relative NAA is

Weight of element 'x' in sample		A_{x^*} in sample $\times (e^{\lambda t})_{sam}$		
Weight of element 'x' in standards	_	A_{r^*} in sample $\times (e^{\lambda t})_{std}$		

Knowing the activities of 'x' in the sample and in standard, the sample and standard decay times and the weight of 'x' in the standard, the weight of element 'x' in the sample can be calculated.

3. Result and discussion

3.1 Quality assurance

Quality control test was performed to investigate the reliability of the analysis by measuring different elements in Certified Reference Material IAEA-SL-1 and Whey powder IAEA-155 relative to IAEA-Soil-7 and comparing the measured values with the certified ones.

3.2 Concentration of As in rice samples

In the present study, the investigations of the elements were performed by means of the Neutron Activation Analysis (NAA). We As in the long irradiation process. The concentrations of As in rice samples are shown in the Table 3 and Figure 2 to 4.

Rice variety	Sample	Concentration of As (mg kg ⁻¹)
BRRI-28	SP-02	BDL
	SP-05	BDL
Γ	SP-08	0.16±0.003
Γ	SP-11	0.2±0.01
Γ	SP-14	0.11±0.01
	SP-17	0.2±0.002
BRRI -29	SP-03	BDL
	SP-06	BDL
	SP-09	0.2±0.003
	SP-12	BDL
	SP-15	BDL
	SP-18	0.17±0.003
Miniket	SP-01	BDL
Ī	SP-04	BDL
	SP-07	0.116±0.002
Ī	SP-10	0.2±0.01
Γ	SP-13	BDL
Ī	SP-16	0.12±0.002

Table 3. Concentrations of As in rice samples determined via NAA.

[BDL=Below Detection Limit]

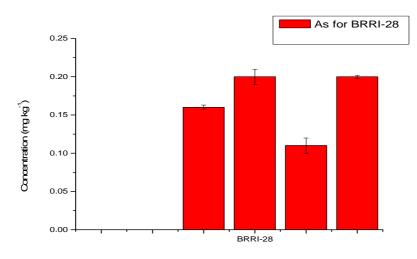


Figure 2. Concentration of As in BRRI-28 variety.

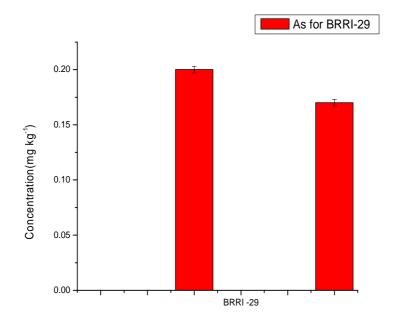


Figure 3. Concentration of As in BRRI-29 variety.

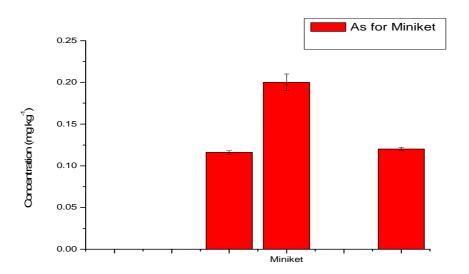


Figure 4. Concentration of As in Miniket variety.

The concentration of arsenic (As) ranging from 0 to 0.2 ± 0.002 mg kg⁻¹ for BRRI-28, 0 to 0.2 ± 0.003 mg kg⁻¹ for BRRI -29 and 0 to 0.2 ± 0.01 mg kg⁻¹ for Miniket. The maximum and minimum average concentration of arsenic (As) was in BRRI-28 and BRRI -29.Rice is a bio-accumulative plant for arsenic (Welna *et al.* 2015). Rice may be significant source of As (Mondal & Polya 2008). Moreover, another important source of arsenic is the use of agrochemicals in paddy field such as a fertilizer, hormone, fungicide, insecticide or soil treatment that improves the production of crop (Kongsria *et al.* 2016). Recently, the JECFA proposed a maximum level of 0.2 mg kg⁻¹ arsenic in rice (FAO/WHO 2014). Due to irrigation with arsenic polluted water higher accumulation of As were informed in Bengal Delta (Meharg & Rahman 2003). In this study the concentration of As found in alarming level for some samples according to maximum permissible level proposed by JECFA. Comparison of arsenic (As) in rice samples from different investigations are shown in Table 4.

Table 4. Comparison of arsenic (As) in rice samples from different investigations around the world. Result express as mean. (Unit: mg kg⁻¹)

Country (reference)	As		
Bangladesh (present study)	0.08		
China(Moon et al. 2012)	0.55		
Indonesia(Moon et al. 2012)	0.08		
Japan(Moon et al. 2012)	0.10		
Korea(Moon et al. 2012)	0.13		
Malaysia(Moon et al. 2012)	0.11		
Philippines(Moon et al. 2012)	0.07		
Thailand(Moon et al. 2012)	0.09		
Japan(Tsukada et al. 2007)	0.085		
Tanzania(Mohammed & Spyrou 2009)	-		

[- indicates not measured]

Officially arsenic (As) is not essential but it has harmful effect to human health. Table 5 indicates side effects both for deficiency and overdose.

Table 5. Side effects of deficiency and overdose for arsenic (As).

Element	Deficiency risk factors	Overdose risk factors		
As	NE	As can cause lung and skin cancers and may cause other cancers (Mondal & Polya 2008).		

[NE= Not establish]

Each adult person in Bangladesh consumes 441 grams of rice a day (Abdullah *et al.* 2005). The values of adequate intake (AI) or recommended dietary intake (RDI) were fixed by the institute of medicine (IOM) of United States (Institue of Medicine, Food and Nutrition Board, 2004). Tolerable daily intake (TDI) for arsenic (As) is shown in Table 6.

Table 6. Tolerable daily intake (TDI) of adult with 70 kg body weight.

Elements	RDI,	TDI	BRRI-28	BRRI-29	Miniket consumers
	mg/day		consumers	consumers	Daily intake(mg/day)
			Daily	Daily	
			intake(mg/day)	intake(mg/day)	
As	NE	150µg/day	.0492	0.0271	0.0320
		(Nedjimi et al.			
		2015)			

[NE= Not establish]

The comparison shows that the Daily Intake of As is less than Tolerable Daily Intake (TDI). The Daily Intake of arsenic (As) is in safe limit.

4. Conclusion and future work

The concentrations of arsenic (As) in three kinds of rice samples were analyzed using Neutron Activation Analysis (NAA). The difference between certified reference values and measured values indicated accuracy of this method. For all varieties of rice samples, average concentration of arsenic (As) was within safe limit. But the concentration of arsenic (As) found in alarming level for some samples according to JECFA. In future other

regions of Bangladesh with other varieties of rice will be analyzed.

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References

Abdullah, A.B., Ito, S. & Adhana, K. (2005), "Estimate of Rice Consumption in Asian Countries and the World towards 2050", Tottori University, Japan, p.28-42.

Ayasekera, R.J. & Freitas M.C. (2005), "Concentration Levels of Major and Trace Elements in Rice from Sri Lanka as Determined by the k0 Standardization Method", *Biological Trace Element Research*, Vol. 103.

Costa, B.E.S., Coelho L.M., Araújo, C.S.T., Rezende, H.C. & Coelho, N.M.M. (2016), "Analytical Strategies for the Determination of Arsenic in Rice", *Journal of Chemistry*, Article ID 1427154, 11 pages.

D'Ilio, S., Alessandrelli, M., Cresti, R., Forte, G. & Caroli, S. (2002), "Arsenic content of various types of rice as determined by plasma- based techniques", *Microchemical Journal*, 73: 195–201.

Diyabalanage, S., Navarathna, T., Abeysundara, H.T.K., Rajapakse, S. & Chandrajith, R. (2016), "Trace elements in native and improved paddy rice from different climatic regions of SriLanka: implications for public health", *SpringerPlus*, 5:1864.

Donisa, C., Mocanu, R., Steinnes, E. & Vasu, A. (2000), "Heavy metal pollution by atmospheric transport in natural soils from the northern part of eastern Carpathians", *Water, Air, & Soil Pollution*, 120: 347–358.

Food and Agriculture Organization/World Health Organization (JECFA), (2014), Joint FAO/WHO Food Standards Committee of the Codex Alimentarius.

Huynh, M.P.T., Carrot, F., Ngoc, S.C.P., Vu, M.D. & Revel, G. (1997), "Determination of rare earth elements in rice by INAA and ICP-MS", *Journal of Radioanalytical and Nuclear Chemistry*, Vol. 217, No. 1, 95-99.

Institue of Medicine, Food and Nutrition Board, (2004), Dietary reference intakes for water, potassium, sodium, chloride and sulfate.National Academy of Sciences, Washington DC.

Kongsria, S., Srinuttrakulb, W., Solab, P. & Busamongkola, A. (2016), "Instrumental neutron activation analysis of selected elements in Thai jasmine rice", *Energy Procedia*, 89: 361 – 365.

Lin, Y.P., Teng, T.P. & Chang, T.K. (2002), "Multivariate analysis of soil heavy metal pollution and landscape pattern in Changhua country in Taiwan", *Landscape and Urban Planning*, 62:19–35.

Mahender, A., Anandan, A., Pradhan, S.K. & Pandit, E. (2016), "Rice grain nutritional traits and their enhancement using relevant genes and QTLs through advanced approaches", *SpringerPlus*, 5:2086.

Meharg, A.A. & Rahman, M.M. (2003), "Arsenic contamination of Bangladesh paddy field soils: implications for rice contribution to arsenic consumption", *Environ. Sci. Technol.*, 37(2):229–234.

Mohammed, N.K. & Spyrou, N.M. (2009), "Trace elemental analysis of rice grown in two regions of Tanzania", *J RadioanalNuclChem*, 281:79–82.

Mondal, D. & Polya D.A. (2008), "Rice is a major exposure route for arsenic in Chakdaha block, Nadia district, West Bengal, India: A probabilistic risk assessment", *Applied Geochemistry*, 23(11), 2987–2998.

Moon, J.H., Ebihara, M., Ni, B.F., Arporn, B., Setyo, P., Theresia, R.M., Wee, B.S., Salim, N.A.Abd. & Pabroa, P.C.B. (2012), "A NAA collaborative study in white rice performed in seven Asian countries", *J RadioanalNuclChem*, 291:217–221.

Nedjimi, B., Beladel, B. & Guit, B. (2015), "Multi-element determination in medicinal Juniper tree (Juniperusphoenicea) by instrumental neutron activation analysis", *Journal of Radiation Research and Applied Sciences*.

Parengama, M., Judprasonga, K., Srianujataa, S., Jittinandanaa, S., Laoharojanaphandb, S. & Busamongkob, A. (2010), "Study of nutrients and toxic minerals in rice and legumes by instrumental neutron activation analysis and graphite furnace atomic absorption spectrophotometry", *Journal of Food Composition and Analysis*, 23:340–345.

Tsukada, H., Hasegawa, H., Takeda, A. & Hisamatsu, S. (2007), "Concentrations of major and trace elements in polished rice and paddy soils collected in Aomori, Japan", *Journal of Radioanalytical and Nuclear Chemistry*, Vol. 273, No.1, 199–203.

Welna, M., Szymczycha-Madeja, A. & Pohl, P. (2015), "Comparison of strategies for sample preparation prior to spectrometric measurements for determination and speciation of arsenic in rice", *Trends in Analytical Chemistry*, 65:122–136.